



## Model calculation of the femtosecond carrier dynamics in Al0.48Ga0.52As

Luís G. C. Rego, Leandro H. F. Andrade, and Carlos H. B. Cruz

Citation: Journal of Applied Physics **76**, 3749 (1994); doi: 10.1063/1.357405 View online: http://dx.doi.org/10.1063/1.357405 View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/76/6?ver=pdfcov Published by the AIP Publishing

## Articles you may be interested in

Surface InP/In0.48Ga0.52P quantum dots: Carrier recombination dynamics and their interaction with fluorescent dyes J. Appl. Phys. **114**, 163510 (2013); 10.1063/1.4827188

Photoluminescence properties of the Al0.48In0.52As/InP interface and the diffusion of carriers thereto J. Appl. Phys. **73**, 7804 (1993); 10.1063/1.353954

Subpicosecond photoresponse of carriers in lowtemperature molecular beam epitaxial In0.52Al0.48As/InP Appl. Phys. Lett. **57**, 1543 (1990); 10.1063/1.103347

Nonlinear optical properties of the electronhole plasma in Al0.52Ga0.48As J. Appl. Phys. **62**, 4187 (1987); 10.1063/1.339087

Calculation of the conduction band discontinuity for Ga0.47In0.53As/Al0.48In0.52As heterojunction J. Appl. Phys. **55**, 3176 (1984); 10.1063/1.333348



This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 143.106.108.153 On: Wed, 26 Nov 2014 17:43:01

# Model calculation of the femtosecond carrier dynamics in Al<sub>0.48</sub>Ga<sub>0.52</sub>As

Luís G. C. Rego, Leandro H. F. Andrade, and Carlos H. B. Cruz Instituto de Física "Gleb Wataghin," Universidade Estadual da Campinas, UNICAMP, 13083-970 Campinas, SP, Brazil

(Received 28 February 1994; accepted for publication 8 June 1994)

We present a model calculation capable of investigating the dynamics of photoexcited carriers in the  $Al_{0.48}Ga_{0.52}As$  indirect gap semiconductor. Nearly resonant excitation at the  $\Gamma$  point produces low excess energy carriers, so that we use Boltzmann like equations and assume thermalized carrier distributions for each of the conduction valleys. Some aspects of the carrier dynamics are discussed and pump and probe measurements are compared to the calculated saturation bleaching, evidencing a very good agreement between theory and experiment. We obtain a value of 3.5 eV/Å for the  $D_{\Gamma X}$  deformation potential.

## I. INTRODUCTION

The determination of intervalley scattering rates in GaAs and associated alloys has been the subject of interest since the '60's, due to the fact that it is one of the main processes of thermal relaxation of hot carriers in semiconductors and, therefore, plays an important role in studies of carrier dynamics and fast devices design. Also the  $Al_xGa_{1-x}As$  alloy has been receiving increasing attention.<sup>1-5</sup> Optical measurements are the best suited for assessing intervalley scattering, since presently only these techniques allow for the necessary time resolution, which is in the subpicosecond range.<sup>6,7</sup> Most of these studies have been done on GaAs,<sup>7-10</sup> using 2.0 eV excitation, but in this case the nonequilibrium carriers are created with high excess energies, complicating the model to be considered and analysis of the data, because many relaxation processes must be taken into account for nonresonant excitation and there are also band anisotropies and nonparabolicities that must be considered for high energy states.

There are various optical studies on the Al<sub>x</sub>Ga<sub>1-x</sub>As alloys in the direct gap region<sup>1,3,11</sup> and also on the indirect gap region;<sup>2,4,5,12</sup> however, despite the amount of theoretical and experimental works, the dynamical behavior of the nonequilibrium carriers is not completely understood for this material, in particular, the intervalley coupling constants envolving the  $\Gamma$  conduction band and the satellite valleys is not known with certainty.

We focus our attention on the  $Al_rGa_{1-r}As$  alloy in the composition range x > 0.45. For an aluminum concentration greater than 43% this material becomes an indirect gap semiconductor with its band gap defined as the difference between the top of the valence band, at the  $\Gamma$  point, and the minimum of the X valley. In this work we model the physical conditions of the experiment made by Andrade et al.<sup>5</sup> where the carriers are optically injected very close to the bottom of the  $\Gamma$  valley by 2.0 eV photons and, for x = 0.48, their excess energy is about 15 meV. For this low excess energy it is possible to assume a very fast carrier thermalization<sup>13</sup> and the conduction and valence bands at the  $\Gamma$  point are adequately described by isotropic effective masses. Due to their high density of states, nonparabolicities in the satellite valleys are not relevant when the density of photoinjected carriers is small.<sup>10</sup> Theoretical calculations are compared with experimental results obtained for an MOCVD (metalorganic

chemical vapor deposition) grown alloy with x=0.48 and show a very good agreement between the experimental and calculated differential transmittance curves.

## **II. THEORETICAL MODEL**

Recent works<sup>9,10</sup> show that nonequilibrium carriers created in the  $\Gamma$  valley of GaAs by 2.0 eV photons, with densities ranging from  $5.10^{16}$  to  $10^{18}$  cm<sup>-3</sup> and initial excess energies of approximately 200 meV up to 560 meV, achieve a quasi-equilibrium regime in time intervals of the order of 50-300 fs. When the quasi-equilibrium is attained this carrier population can be conveniently described in terms of its quasi-temperature and density, so that its time evolution is calculated by some coupled equations. Other works<sup>13,14</sup> show that, as a result of the same sort of excitation in GaAs,  $\Gamma$ valley carriers have to be considered as thermalized in order to explain the observed experimental results that occur within 1 ps time interval. As indicated by these previous results and taking into account that we are considering photocreated pairs with a much lower excess energy (around 15 meV), we argue that the quasi-equilibrium approximation can be conveniently used to describe our physical picture. However, carriers having very different wave vectors are weakly coupled by the Coulomb potential, since it presents a  $q^{-2}$  dependence in its matrix element  $(q = |\mathbf{k} - \mathbf{k}'|)$ . Taking into account this fact, we assume that each set of satellite valleys (4L and 3X), and the energy bands at the center of the Brillouin zone (conduction and valence bands), are characterized by their own quasi-temperatures. All the X electrons are assumed to have a quasi-temperature  $T^X$ , the L electrons have a quasi-temperature  $T^L$ , and the carriers with wave vectors belonging to the center of the k space,  $\Gamma$  electrons and holes, have a quasi-temperature  $T^{\Gamma}$ .

With these assumptions we define four carrier populations that are represented by distinct distribution functions of type

$$f_{\mathbf{k}}[T_{i}(t),\mu_{i}(t)] = \frac{1}{\exp\{\beta_{i}(t)[E_{i}(k)-\mu_{i}(t)]\}+1},$$
 (1)

where the index i is used to distinguish the quasi-tempera-

tures and quasi-chemical potentials of these four populations. Their time evolution is determined by the following set of coupled equations

$$\frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} = \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} \bigg|_{\text{Opt}} + \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} \bigg|_{\Gamma X} + \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} \bigg|_{\Gamma L} + \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} \bigg|_{\text{Fr}} + \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} \bigg|_{\text{Fr}},$$

$$(2a)$$

$$\frac{\partial f_{\mathbf{k}}^{\Gamma,h}}{\partial t} = \frac{\partial f_{\mathbf{k}}^{\Gamma,h}}{\partial t} \bigg|_{\text{Opt}} + \frac{\partial f_{\mathbf{k}}^{\Gamma,h}}{\partial t} \bigg|_{\text{Fr}}, \qquad (2b)$$

$$\frac{\partial f_{\mathbf{k}}^{X}}{\partial t} = \frac{\partial f_{\mathbf{k}}^{X}}{\partial t}\Big|_{\Gamma X} + \frac{\partial f_{\mathbf{k}}^{X}}{\partial t}\Big|_{L X} + \frac{\partial f_{\mathbf{k}}^{X}}{\partial t}\Big|_{Fr} + \frac{\partial f_{\mathbf{k}}^{X}}{\partial t}\Big|_{Alloy}, \qquad (2c)$$

$$\frac{\partial f_{\mathbf{k}}^{L}}{\partial t} = \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} \bigg|_{\Gamma L} + \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} \bigg|_{LX} + \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} \bigg|_{Fr} + \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} \bigg|_{Alloy}, \qquad (2d)$$

where the superscripts  $(\Gamma, e)$ ,  $(\Gamma, h)$ , (X), and (L) represent the  $\Gamma$  electrons, the holes, the X valley electrons, and the L valley electrons, respectively. Opt component describes the carrier injection, through direct absorption, by the optical pulse;  $\Gamma X$ ,  $\Gamma L$ , and LX indicate the phonon assisted intervalley coupling among  $\Gamma$ , X, and L valleys via intervalley deformation potential; the Fr term denotes the intraband interaction of carriers and LO phonons due to the screened Fröhlich potential and, finally, Alloy takes into account the elastic scattering of carriers by alloy potential fluctuations (or zero-phonon transitions).

We assume that the phonon populations are in equilibrium, described by a Planckian distribution with a temperature that is equal to lattice temperature ( $T_L = 300$  K), because we create a low density nonequilibrium carrier population (about  $3.10^{16}$  cm<sup>-3</sup>) that is not able to disturb the phonon populations far away from its equilibrium state. Following Adachi<sup>15</sup> we describe the two LO phonons dispersion curves by a single effective one:  $\omega_{10} \equiv 0.52(\omega_{10}^{GaAs}) + 0.48(\omega_{10}^{AsAl})$ , that takes into account GaAs-like and AlAs-like phonons for this aluminum concentration. The zone center phonons, that participate in the intravalley processes, have energy  $\hbar \omega_{ZC} = 40.5$  meV and the zone edge phonons, that are related to intervalley scatterings, have  $\hbar \omega_{ZE} = 33$  meV.<sup>4</sup>

Each of the interaction terms introduced in (1) (except the one due to alloy potential fluctuations, that we treat by the relaxation time approximation) is represented by rate equations of the form<sup>16</sup>

$$\frac{\partial f_{\mathbf{k}}(t)}{\partial t}\bigg|_{\mathrm{INT}} = \sum_{\mathbf{k}'} \{f_{\mathbf{k}'}(t) [1 - f_{\mathbf{k}}(t)] W(\mathbf{k}', \mathbf{k}) - f_{\mathbf{k}}(t) [1 - f_{\mathbf{k}'}(t)] W(\mathbf{k}, \mathbf{k}')\},$$
(3)

where the subscript INT represents each one of the interactions of Eqs. (2) and W is the quantum transition rate, given by Fermi's golden rule and averaged over the phonon (or photon) ensemble

#### 3750 J. Appl. Phys., Vol. 76, No. 6, 15 September 1994

$$W(\mathbf{k},\mathbf{k}\pm\mathbf{q}) = \frac{2\pi}{\hbar} \sum_{\mathbf{q}} |\langle \mathbf{k}\pm\mathbf{q}|H'|\mathbf{k}\rangle|^2 [\nu\delta(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}) - \hbar w) + (\nu+1)\delta(E_{\mathbf{k}-\mathbf{q}}-E_{\mathbf{k}}+\hbar w)], \quad (4)$$

so that  $\nu$  stands for the phonon or photon populations, depending on the interaction considered, and  $\langle \mathbf{k} \pm \mathbf{q} | H' | \mathbf{k} \rangle$  its matrix element.

For the intervalley scattering terms we write<sup>17</sup>

$$|\langle \mathbf{k} \pm \mathbf{q} | H' | \mathbf{k} \rangle|^2 = \frac{(D_{i,j})^2 \hbar}{2 V \rho \omega_{ZE}}, \qquad (5)$$

where  $D_{i,j}$  is an effective deformation potential coupling constant (*i* and *j* representing  $\Gamma$ , *X*, and *L*), *V* is the volume of the sample,  $\rho$  its density, and  $\omega_{ZE}$  the zone edge effective phonon frequency.

The matrix element of the screened Fröhlich potential is<sup>18</sup>

$$|\langle \mathbf{k} \pm \mathbf{q} | H' | \mathbf{k} \rangle|^2 = \frac{2 \pi e^2 \hbar w_{\text{ZC}}}{V} \left( \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_0} \right) \frac{1}{q^2 \epsilon^2}, \qquad (6)$$

such that  $\epsilon_{\infty}$  and  $\epsilon_0$  are the high and low frequency dielectric constants, respectively, q is the modulus of the LO phonon wave vector  $(q=|\mathbf{k}'-\mathbf{k}|)$ , and  $\epsilon$  is the dielectric constant in the static limit of the random phase approximation.

Intervalley scattering caused by potential fluctuations in the  $Al_xGa_{1-x}As$  alloy is treated in Refs. 12 and 19. Different from the phonon induced interactions that scatter the carriers due to periodic potential fluctuations of the ions and the band structure, the alloys present random fluctuations of potential that break the translation symmetry, inducing transitions without the intervention of phonons (zero-phonon). A precise treatment of this problem is difficult and the virtual crystal model is usually used (Grein *et al.*<sup>19</sup> compare different approaches to treat the problem). This process is expected to be weaker than the phonon scattering contributions,<sup>12</sup> so we describe alloy scattering by means of the relaxation time approximation

$$\left. \frac{\partial f_{\mathbf{k}}(t)}{\partial t} \right|_{\text{Alloy}} = -\frac{f_{\mathbf{k}}(t) - f_{\mathbf{k}}(\text{equil})}{\tau_{i}} , \qquad (7)$$

where  $f_k(\text{equil})$  is the Fermi–Dirac distribution function at the equilibrium and the relaxation time  $\tau$  is written following Kalt *et al.*<sup>12</sup> as

$$\tau_i^{-1} = \frac{a^3 x (1-x) (V_{\Gamma i}^{\text{Alloy}})^2 m_i^{3/2} \Delta_{\Gamma i}^{1/2}}{2^{3/2} \pi \hbar^4} , \qquad (8)$$

where x is the aluminum concentration in the sample, a the lattice parameter,  $\Delta_{\Gamma i}$  the energy difference between the extremes of the  $\Gamma$  valley and the valley designated by *i*,  $m_i$  is the effective mass of the electron in either the X or L valleys, and  $V_{\Gamma,i}^{\text{Alloy}}$  is the coupling potential for alloy potential fluctuations. Following Kalt *et al.*<sup>12</sup> we consider  $V_{\Gamma,X}^{\text{Alloy}} = 110$  meV and assume that  $V_{\Gamma,L}^{\text{Alloy}} = V_{\Gamma,X}^{\text{Alloy}}$ , to obtain  $\tau_X = 1$  ps and  $\tau_L = 2.2$  ps.

In order to obtain the time evolution of the density of photoinjected carriers  $(N \equiv 1/V \Sigma_k f_k)$ , and also their energy density  $(U \equiv 1/V \Sigma_k E_k f_k)$ , we make use of Eqs. (2) to write

Rego et al.

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] 143 106 108 153 Op; Wed, 26 Nov 2014 17:43:01

$$\dot{U}^{\Gamma} = \frac{1}{V} \sum_{\mathbf{k}} \left( E_{\mathbf{k}}^{\Gamma,e} \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} + E_{\mathbf{k}}^{\Gamma,b} \frac{\partial f_{\mathbf{k}}^{\Gamma,b}}{\partial t} \right)$$
$$= \dot{U}_{\text{Opt}}^{\Gamma} + \dot{U}_{\Gamma X}^{\Gamma} + \dot{U}_{\Gamma L}^{\Gamma} + \dot{U}_{\text{Fr}}^{\Gamma} + \dot{U}_{\text{Alloy}}^{\Gamma}, \qquad (9a)$$

$$\dot{N}^{\Gamma,e} = \frac{1}{V} \sum_{\mathbf{k}} \frac{\partial f_{\mathbf{k}}^{\Gamma,e}}{\partial t} = \dot{N}_{\text{Opt}}^{\Gamma,e} + \dot{N}_{\Gamma X}^{\Gamma,e} + \dot{N}_{\Gamma L}^{\Gamma,e} + \dot{N}_{\text{Alloy}}^{\Gamma,e}, \quad (9b)$$

$$\dot{N}^{\Gamma,h} = \frac{1}{V} \sum_{\mathbf{k}} \frac{\partial f_{\mathbf{k}}^{\Gamma,h}}{\partial t} = \dot{N}_{\text{Opt}}^{\Gamma,h}, \qquad (9c)$$

$$\dot{U}^{X} = \frac{1}{V} \sum_{\mathbf{k}} E_{\mathbf{k}}^{X} \frac{\partial f_{\mathbf{k}}^{X}}{\partial t} = \dot{U}_{\Gamma X}^{X} + \dot{U}_{L X}^{X} + \dot{U}_{Fr}^{X} + \dot{U}_{Alloy}^{X}, \quad (9d)$$

$$\dot{N}^{X} = \frac{1}{V} \sum_{\mathbf{k}} \frac{\partial f_{\mathbf{k}}^{X}}{\partial t} = \dot{N}_{\Gamma X}^{X} + \dot{N}_{L X}^{X} + \dot{N}_{\text{Alloy}}^{X}, \qquad (9e)$$

$$\dot{U}^{L} = \frac{1}{V} \sum_{\mathbf{k}} E_{\mathbf{k}}^{L} \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} = \dot{U}_{\Gamma L}^{L} + \dot{U}_{LX}^{L} + \dot{U}_{Fr}^{L} + \dot{U}_{Alloy}^{L}, \qquad (9f)$$

$$\dot{N}^{L} = \frac{1}{V} \sum_{\mathbf{k}} \frac{\partial f_{\mathbf{k}}^{L}}{\partial t} = \dot{N}_{\Gamma L}^{L} + \dot{N}_{LX}^{L} + \dot{N}_{Alloy}^{L}, \qquad (9g)$$

where  $E_{\mathbf{k}}$  is the energy of the electronic state  $\mathbf{k}$ .

Because of the threefold degenerace of the X valleys and the fourfold degenerace of the L valleys we have

$$\dot{N}_{\Gamma X}^{\Gamma} = -3\dot{N}_{\Gamma X}^{X},\tag{10}$$

$$\dot{N}_{\Gamma L}^{\Gamma} = -4\dot{N}_{\Gamma L}^{L}, \qquad (11)$$

$$4\dot{N}_{LX}^{L} = -3\dot{N}_{LX}^{X},$$
 (12)

$$\dot{N}_{\text{Alloy}}^{\Gamma} = -3\dot{N}_{\text{Alloy}}^{X} - 4\dot{N}_{\text{Alloy}}^{L}.$$
(13)

The relation among the kinetic Eqs. (9) and the quasitemperatures and quasi-chemical potentials can be found by differentiating U and N with respect to time, where

$$U(t) = \frac{1}{V} \sum_{\mathbf{k}} E_{\mathbf{k}} \left[ \exp \left( \frac{1}{kT(t)} \left[ E_{\mathbf{k}} - \mu(t) \right] \right) + 1 \right]^{-1}, \quad (14)$$

$$N(t) = \frac{1}{V} \sum_{\mathbf{k}} \left[ \exp\left(\frac{1}{kT(t)} \left[E_{\mathbf{k}} - \mu(t)\right]\right) + 1 \right]^{-1}.$$
 (15)

In doing this we obtain a set of coupled differential equations to the intensive parameters that is numerically integrated to give their temporal evolution.

### **III. RESULTS AND COMMENTS**

In order to establish a connection between the theoretical model and experimental data we reproduced the measured differential transmittance recovery curve obtained in the experiments of Andrade *et al.*<sup>5</sup> (Fig. 1). The main feature in the data is a sudden raise of the transmittance coefficient, due to electron-hole pair generation at the  $\Gamma$  point, and its subsequent-ultrafast (~120 fs time constant) decay, caused by intervalley scattering of  $\Gamma$  electrons to satellite valleys. The pump pulse has a sech<sup>2</sup> profile with 90 fs full width at half maximum (FWHM) and its spectral distribution is ob-



FIG. 1. Theoretical fitting of the experimental curve obtained by Andrade *et al.* (see Ref. 5). The dashed line represents the calculated values and the solid line the experimental data. It is considered  $D_{\Gamma X}=3.5$  eV/Å,  $D_{\Gamma L}=5.0$  eV/Å, and  $D_{LX}=2.4$  eV/Å.

tained through a Fourier transform. The fitting of the experimental data at delay times longer than 700 fs does not depend on  $D_{\Gamma X}$  or  $D_{\Gamma L}$ , being affected mostly by the value of the excess energy of the photocreated pairs. A good agreement between the curves at long delay times is obtained only for an excess energy of 15 meV, in accordance with the experiment.

Because alloy potential fluctuation scatterings are characterized by time constants of the order of picoseconds, the transfer rate of zone center electrons to satellite valleys is mainly determined by nonpolar optical phonon interaction. Furthermore, XL scattering is a very indirect mechanism to be realized by the data of Fig. 1, so we consider the results of Güncer and Ferry,<sup>20</sup> and use  $D_{XL}$ =2.4 eV/Å (for an effective phonon dispersion relation), obtained for the Al<sub>0.6</sub>Ga<sub>0.4</sub>As alloy.

It was observed that a set of  $D_{\Gamma X}$  and  $D_{\Gamma L}$  pairs obeying the relation  $\alpha (D_{\Gamma X})^2 + \beta (D_{\Gamma L})^2 = 1$ , where  $\alpha = 0.018$  and  $\beta$ =0.044, gives the best agreement between the experimental and theoretical data. Ulman et al.<sup>11</sup> obtained  $D_{\Gamma L} = 5.0 \text{ eV/Å}$ for the Al<sub>0.1</sub>Ga<sub>0.9</sub>As alloy through an experiment capable of measuring only  $\Gamma L$  transitions. This value corresponds to an effective deformation potential that takes into account different types of phonons. Considering the works of Zollner et al.<sup>21,22</sup> we conclude that this value must be due essentially to LA and LO phonons. It is also interesting to notice in Ref. 22 that the sum of the intervalley deformation potential contributions due to LO and LA phonons is weakly dependent on k for a wide range of electron energies. Using  $D_{\Gamma L} = 5.0$ eV/Å we obtain for the  $\Gamma X$  intervalley transition  $D_{\Gamma X} = 3.5$ eV/Å. The curves of Fig. 1 show very good agreement between theory and experiment for  $D_{\Gamma X}$ =3.5 eV/Å,  $D_{\Gamma L}$ =5.0 eV/Å, and  $D_{XL}$ =2.4 eV/Å. The sensitivity of this fit upon changes on  $D_{\Gamma X}$  can be seen in Fig. 2, where we considered  $D_{\Gamma X}$  equal to 2.0 eV/Å and 5.0 eV/Å, maintaining the above values for  $D_{\Gamma L}$  and  $D_{XL}$ . As we increase the intervalley coupling, the photoinjected carriers leave the  $\Gamma$  valley quicker and the recovery of the saturation bleaching becomes faster. Taking into account the  $\Gamma X$  intervalley deformation poten-

J. Appl. Phys., Vol. 76, No. 6, 15 September 1994

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] II 143.106.108.153 On: Wed. 26 Nov 2014 17:43:01



FIG. 2. Sensitivity of the fitting upon changes on  $D_{\Gamma X}$ . It is considered  $D_{\Gamma L}=5.0 \text{ eV}/\text{\AA}$  and  $D_{L X}=2.4 \text{ eV}/\text{\AA}$ ,  $D_{\Gamma X}$  is 2.0 eV/Å (dotted) and 5.0 eV/Å (dashed). The solid line curve represents the experimental data.

tials calculated by Zollner<sup>22</sup> for GaAs, we observe that for  $\Gamma$  electrons with very small energies, as is the case in this work where  $|\mathbf{k}|$  is centered around  $2.5 \times 10^6$  cm<sup>-1</sup>, the effective deformation potential we obtain corresponds to the sum of contributions ascribed to LO phonons ( $D_{\Gamma X, LO} \approx 0.75 \times D_{\Gamma X} \approx 2.6 \text{ eV/Å}$ ) and TO+TA phonons ( $D_{\Gamma X, T} \approx 0.25 \times D_{\Gamma X} \approx 0.9 \text{ eV/Å}$ ), revealing a close agreement with the values of Zollner.

From the model calculations, it is possible to obtain further insight into the carrier thermodynamical properties. First we consider the carrier densities, as shown in Fig. 3. We observe that the density of holes increases with the pump pulse excitation and remains constant after it has finished. However, the density of  $\Gamma$  electrons grows until it reaches a maximum value, lower than the hole concentration, that corresponds to the instant when the rate of carrier generation (due to direct absorption) is the same as the intervalley scattering rates. From this time on the density of  $\Gamma$  electrons decreases and the satellite valleys begin to populate. The intervalley transfer of electrons is directly associated with the



FIG. 3. Time evolution of the carrier densities as a function of delay time. Solid line represents the holes, dotted line the  $\Gamma$  electrons, dot-dashed line the 3 X valleys and long-dashed line the 4L valleys.

3752 J. Appl. Phys., Vol. 76, No. 6, 15 September 1994



FIG. 4. Carrier quasi-temperatures as a function of delay time for  $\Gamma$  electrons and holes (solid line), L electrons (dotted line), and X electrons (dashed line).

time decay of the transmittance curve, as we can see from Figs. 1 and 3.

Figure 4 shows the carrier quasi-temperatures as a function of delay time. Their initial values correspond to a condition in which the system is in thermal equilibrium at room temperature. An interesting feature is that the quasitemperature of the carriers having wave vectors around the center of the Brillouin zone decreases, while the pump pulse strongly interacts with the sample and, afterward, increases until it attains room temperature. The initial decrease can be understood since the pump photon energy creates electronhole pairs with an excess energy of 15 meV, smaller than the lattice thermal energy at room temperature. It is possible to estimate the quasi-temperature of the created electron-hole pairs using the equipartion law, for a pair with 15 meV excess energy we have  $T \approx \epsilon/3k_B = 58$  K, where  $k_B$  is the Boltzmann constant. Effectively, the minimum temperature observed in Fig. 4 is higher than this ( $\simeq 100$  K), because the carriers are also heated due to the absorption of LO phonons via intravalley polar collisions. On the other hand, we observe the opposite behavior for the satellite valleys, their quasi-temperatures raise and then decrease towards room temperature. This kind of behavior is understood when we realize that, due to the  $\Gamma$  to X (or L) transition, part of the  $\Gamma$ electron potential energy is transformed into kinetic energy. For a 48% aluminum concentration sample the difference in energy between the  $\Gamma$  and X minima is 96 meV and between  $\Gamma$  and L, 43 meV.<sup>15</sup> We can verify that the kinetic energy associated with the maximum of the quasi-temperature curves correspond approximately to the difference in energy between  $\Gamma$  and X (or L) valleys, minus the zone edge phonon energy. This argument is able to explain the higher values of the X quasi-temperature, when compared with those of the Lelectrons. After the peak values are reached the quasitemperatures decrease due to thermalization with the lattice by means of intravalley polar collisions with the LO phonons; the rate of decay of the X quasi-temperature curve is greater than the one of L because the density of states of a single X valley is much bigger than that of L, and this multiplies the possibility of collisions.

Rego et al.

#### **IV. CONCLUSIONS**

A model calculation was developed to describe the generation, and subsequent behavior, of the nearly resonant photoexcited carriers in the Al<sub>0.48</sub>Ga<sub>0.52</sub>As alloy, with indirect band gap. The occupation rates of the satellite valleys and the carrier quasi-temperatures were calculated as a function of time. We get very good agreement between theoretical and experimental results, so that we obtain an effective  $\Gamma X$  coupling constant, due to nonpolar optical interaction,  $D_{\Gamma X}=3.5$ eV/Å. For the  $\Gamma L$  scatterings we assume  $D_{\Gamma L}=5.0$  eV/Å, following Ulman *et al.*<sup>11</sup> We believe that these ultrafast optical nonlinearities (~120 fs) make the Al<sub>x</sub>Ga<sub>1-x</sub>As, at the indirect gap region, an interesting material from the point of view of ultrafast recovery processes.

### ACKNOWLEDGMENTS

We acknowledge financial support from the Brazilian Research Agencies: CNPq and FAPESP.

<sup>1</sup>A. J. Taylor, D. J. Erskine, and C. L. Tang, J. Opt. Soc. Am. 2, 663 (1985).
 <sup>2</sup>H. Kalt, W. W. Rühle, and K. Reimann, Solid-State Electron. 32, 1819 (1989).

<sup>3</sup>D. S. Kim, J. M. Jacob, J. F. Zhou, J. J. Song, H. Hou, C. W. Tu, and H. Morkoç, Phys. Rev. B **45**, 13973 (1992).

- <sup>4</sup>W. B. Wang, K. Shum, R. R. Alfano, D. Szmyd, and A. J. Nozik, Phys. Rev. Lett. **68**, 662 (1992).
- <sup>5</sup>L. H. F. Andrade, M. A. Sacilotti, and C. H. Brito Cruz, Solid State Commun. **85**, 953 (1993).
- <sup>6</sup>W. Z. Lin, R. W. Schoenlein, J. Fujimoto, and E. P. Ippen, IEEE J. Quantum Electron. **12**, 267 (1988).
- <sup>7</sup>P. C. Becker, H. L. Fragnito, C. H. Brito Cruz, J. Shah, R. L. Fork, J. E. Cuninghan, J. E. Henry, and C. V. Shank, Appl. Phys. Lett. **53**, 2089 (1988).
- <sup>8</sup>C. L. Collins and P. Yu, Phys. Rev. B **30**, 4501 (1984).
- <sup>9</sup>L. Rota, P. Lugli, T. Elsaesser, and J. Shah, Phys. Rev. B 47, 4226 (1993).
   <sup>10</sup>U. Hohenester, P. Supanicic, P. Kocevar, X. Q. Zhou, W. Kütt, and H. Kurz, Phys. Rev. B 47, 13233 (1993).
- <sup>11</sup> M. Ulman, D. W. Bailey, L. H. Acioli, F. G. Vallée, C. J. Stanton, E. P. Ippen, and J. G. Fujimoto, Phys. Rev. B 47, 10267 (1993).
- <sup>12</sup> H. Kalt, W. W. Rúhle, K. Reimann, M. Rinker, and E. Bauser, Phys. Rev. B 43, 12364 (1991).
- <sup>13</sup> T. Gong, P. M. Fauchet, J. Young, and P. J. Kelly, Phys. Rev. B 44, 6542 (1991).
- <sup>14</sup>D. S. Kim and P. Y. Yu, Phys. Rev. B 43, 4158 (1991).
- <sup>15</sup>S. Adachi, J. Appl. Phys. 58, 1 (1985).
- <sup>16</sup> B. R. Nag, *Electron Transport in Semiconductors*, Springer Series in Solid State Science, Vol. 11 (Springer, Berlin, 1980).
- <sup>17</sup>E. M. Conwell, *High Field Transport in Semiconductors*, Solid State Physics Suppl., Vol. 9 (Academic, New York, 1968).
- <sup>18</sup>J. Collet and T. Amand, J. Phys. Chem. Solids 47, 153 (1986).
- <sup>19</sup>C. H. Grein, S. Zollner, and M. Cardona, Phys. Rev. B 44, 12761 (1991).
- <sup>20</sup>S. E. Güncer and D. K. Ferry, Phys. Rev. B 46, 15309 (1992).
- <sup>21</sup>S. Zollner, S. Gopalan, and M. Cardona, Appl. Phys. Lett. 54, 614 (1989).
- <sup>22</sup>S. Zollner, S. Gopalan, and M. Cardona, J. Appl. Phys. 68, 1682 (1990).

<sup>[</sup>This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] 143 106 108 153 On: Wed. 26 Nov 2014 17:43:01